Interactions in Confined Polariton Condensates

Lydie Ferrier,[1] Esther Wertz,[1] Robert Johne,[2] Dmitry D. Solnyshkov,[2] Pascale Senellart,[1] Isabelle Sagnes,[1] Aristide Lemaître,[1] Guillaume Malpuech,[2] and Jacqueline Bloch[1,*]

[1] *CNRS–Laboratoire de Photonique et Nanostructures, Route de Nozay, 91460 Marcoussis, France*
[2] *LASMEA, CNRS, University Blaise Pascal, 24 avenue des Landais, 63177 Aubière cedex, France*
(Received 11 December 2010; revised manuscript received 1 February 2011; published 21 March 2011)

We investigate the effect of interactions in zero-dimensional polariton condensates. The shape of the condensate wave function is shown to be modified by repulsive interactions with the reservoir of uncondensed excitons. In large micropillar cavities, when uncondensed excitons are located at the center, the condensate is ejected toward the pillar edges. The same effect results in the generation of optical traps in wire cavities. Once polariton condensates are spatially separated from the excitonic reservoir, spectral signatures of polariton-polariton interactions within the condensate are evidenced.

DOI: 10.1103/PhysRevLett.106.126401 PACS numbers: 71.36.+c, 42.65.Pc, 73.50.Pz, 78.55.Cr

After the experimental demonstration of Bose-Einstein condensation with cold atoms in 1995 [1,2], a lot of interest has been drawn to the influence of interactions on the physics of atomic Bose-Einstein condensates [3]. Renormalization of the excitation spectra into linear Bogoliubov dispersions has been shown [4], leading to a superfluid behavior of atomic condensates [5]. Interactions also strongly affect finite size condensates: in the presence of repulsive interactions, minimization of the condensate energy leads to a significant spatial expansion of its wave function, which tends to a Thomas-Fermi profile [6,7].

Recently, semiconductor microcavities have appeared as a model system to investigate Bose condensates in a solid state system [8–15]. Their optical properties in the low density regime are governed by bosonic exciton-photon mixed states named cavity polaritons. Despite the two dimensionality of this system, polariton dynamical condensation can occur, giving rise to the formation of coherent light-matter quasicondensates. From their photonic nature, cavity polaritons inherit a light effective mass (10^{-8} that of atoms) allowing condensation effects at high temperatures. Because of their excitonic nature, polaritons interact with phonons [16], excitons, or polaritons [17,18]. One signature of the interactions undergone by polariton condensates is the continuous blueshift of the emission above condensation threshold, discussed by several groups [8,9,13]. Recently, other experimental signatures of these interactions have been studied, such as the renormalization of the polariton dispersion into a linear Bogoliubov profile [19,20] or superfluid behavior [20,21]. A difficulty in the investigation of polariton interactions when nonresonant excitation is considered is to distinguish interaction with the reservoir of uncondensed excitons from polariton-polariton interactions within the condensate.

In this Letter, we investigate interactions in 0D polariton condensates generated under nonresonant optical excitation. Using two resonator geometries we identify the respective role of interactions with the excitonic reservoir and of polariton-polariton interactions within the condensate. Condensation is first investigated in micropillars with a lateral size much larger than the excitation spot diameter. The excitonic reservoir induces a local potential maximum at the pillar center. These repulsive interactions are responsible for the expulsion of the condensate wave function from the excitation area. The second investigated geometry is a microwire excited close to its edge [14]. Repulsive interaction with the excitonic reservoir is responsible for condensation in the optical trap formed between the excitation area and the wire end. Once the condensate is spatially separated from the excitonic reservoir (both in micropillars and in optical traps), self-interactions within the condensate is revealed. A threshold-like behavior in the spectral blueshift of the condensate emission is attributed to polariton-polariton interactions when the mean number of polaritons in the condensate exceeds a few hundred. Finally, we compare these polaritonic features with regular photon lasing obtained at higher excitation power in micropillars [13]. Remarkably, in the photon lasing regime, an opposite spatial behavior is observed: lasing is triggered in the region from which polariton condensates are expelled.

The cavity sample is described in detail in Ref. [13]. Square micropillars (with sizes ranging from 20 to 2 μm) and 200 μm long microwires (with lateral sections ranging from 2 to 4 μm) were fabricated using electron beam lithography and reactive ion etching. Microphotoluminescence experiments are performed on single micropillars and microwires using a single mode cw Ti:sapphire laser focused onto a 2 μm diameter spot with a microscope objective. The sample is maintained at 10 K and the excitation laser energy is tuned typically 100 meV above the lower polariton resonance. The emission is collected through the same objective and imaged on the entrance slit of a monochromator (for microwire measurements, the wire image is parallel to the slit). The spectrally dispersed emission is detected with a nitrogen cooled CCD camera. Local heating of the micropillars is reduced by chopping the laser beam with an acousto-optic modulator with 1% duty cycle at 10 kHz. The lowest cavity mode is chosen detuned by roughly -2 meV with respect to the excitonic resonance.

Microcavity polaritons in high quality factor samples like the present one have been shown to undergo bosonic stimulation under nonresonant excitation. Polariton lasing (also called polariton condensation) has been demonstrated in micropillar [13] and microwire [14] cavities.

Under cw nonresonant pumping, steady-state polariton condensates are generated together with uncondensed higher energy excitons forming an excitonic reservoir. The wave function $\psi(r)$ of polaritons obeys the following mean-field Gross-Pitaevskii equation [22] where a contact interaction between polaritons is considered:

$$\left\{-\frac{\hbar^2}{2m}\nabla^2 + V(r) + g|\psi(r)|^2\right\}\psi(r) = \mu\psi(r), \quad (1)$$

where m is the polariton effective mass, μ its chemical potential, and g the polariton-polariton interaction constant [23,24]. $V(r)$ is the external potential felt by polaritons:

$$V(r) = V_{\mathrm{conf}}(r) + V_{\mathrm{res}}(r). \quad (2)$$

$V_{\mathrm{conf}}(r)$ corresponds to the confinement potential defined by the etched microstructure under study. It will be taken as infinite outside the micropillar or the microwire and equal to zero inside. $V_{\mathrm{res}}(r) = g_{\mathrm{res}}n_{\mathrm{res}}(r)$ is the power-dependent potential resulting from repulsive interactions (described by the interaction constant g_{res}) with uncondensed excitons injected in the excitation area. $n_{\mathrm{res}}(r)$ describes the spatial distribution of uncondensed excitons. Because of the small diffusion coefficient of excitons, the shape of the exciton cloud is given by that of the pump profile. Both interaction terms, V_{res} and $g|\psi(r)|^2$, give rise to a blueshift of the condensate emission and can induce spatial modifications of its wave function [25]. As will be shown in the following, the experimental spatial distributions $|\psi(r)|^2$ and $n_{\mathrm{res}}(r)$ can strongly differ when the excitation spot size is small compared to the cavity lateral size. This allows us to clearly distinguish the effect of each of these interaction terms.

We first investigate the wave function of 0D polaritons generated in square micropillars excited at their center. As shown in Fig. 1(a), spatially resolved photoluminescence performed on a single micropillar indicates the formation of discrete confined polariton states. Figure 1(b) shows emission spectra measured on a single 10 μm square micropillar for different excitation powers. A marked threshold is evidenced in the emission intensity above which polariton condensation is obtained. Notice that at threshold, the emission is strongly multimode. However, for higher excitation powers, mode competition favors condensation on the lowest energy polariton state (see also far field images in Fig. 2 in the supplemental material [26]). Figures 1(c)–1(e) show the spatial distribution of the total emission intensity measured on this micropillar for three excitation powers. Below threshold, the emission presents a Gaussian shape centered on the pillar. As the

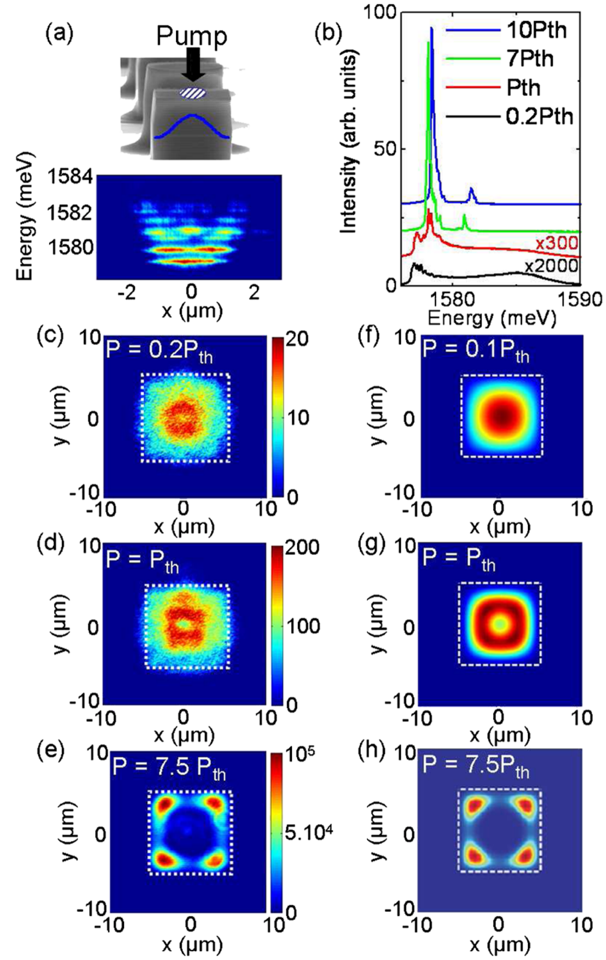

FIG. 1 (color online). (a) Top: Scanning electron microscopy image of a micropillar. The dashed circle and blue line schematically indicate the spatial location of the excitonic reservoir and of confined polaritons; bottom: spatially and spectrally resolved emission of a single 5 μm micropillar measured below threshold. (b) Emission spectra measured on a 10 μm square pillar for several excitation powers. (c)–(e) Spatial distribution of the total emission intensity measured on the same 10 μm pillar for three excitation powers. (f)–(h) Corresponding calculated intensity distributions of the lowest energy polariton state. White dashed squares show the pillar edge.

excitation power is increased, a hole in the intensity distribution appears at the center. Far above threshold, the condensate wave function presents four lobes located in the corners of the pillar, whereas it is vanishing in the center. Figures 1(f)–1(h) show the calculated wave function of the lowest energy polariton state solving Eq. (1), taking into account the interactions within the condensate and including a Gaussian reservoir potential with a 2 μm spatial width and a height proportional to the excitation power. The shape of the emission pattern is perfectly reproduced considering this repulsive interaction with the excitonic reservoir, which pushes away the condensate from the pillar center toward the corners.

Notice that when the excitation spot is not centered on the micropillar, we observe expulsion of the condensate wave function out of the excitation area with asymmetric emission pattern (see Fig. 4 in the supplemental material [26]). Also interesting spatial changes have been obtained when exciting coupled micropillars (these results will be published elsewhere). All these measurements are fully consistent with the interpretation in terms of repulsive interactions with the cloud of uncondensed excitons.

We now consider the generation of 0D polariton condensates in 1D cavities. We have recently shown that the excitonic repulsive potential can be used to define an optically controlled trap when using a single wire cavity [14]. An example of such a trap is shown in Fig. 2(a) where

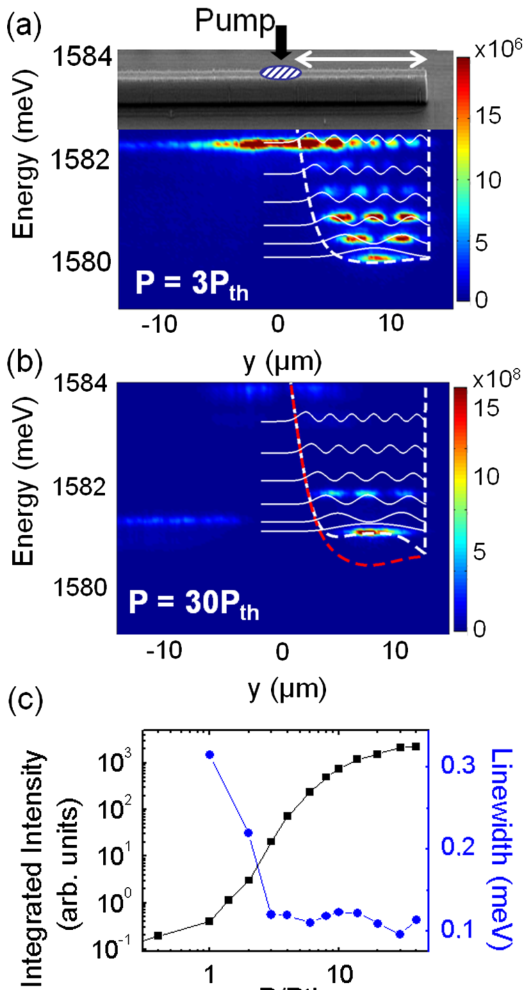

FIG. 2 (color online). (a) Spatially and spectrally resolved emission of a microwire excited 15 μm from its end for $P = 3P_{\rm th}$. White lines: Calculated polariton states confined by the potential shown in white dashed line which includes interaction with the excitonic reservoir. Inset: Scanning electron micrography of the microwire with schematic representation of excitonic reservoir in dashed blue. (b) Same as (a) for $P = 30P_{\rm th}$. The potential in white (red) dashed line also includes (does not include) the potential induced by the polarization occupancy. (c) Integrated intensity and spectral linewidth of the lowest energy polariton state measured in the optical trap as a function of excitation power.

a wire cavity with a 2.5 μm lateral size is excited 15 μm away from its end. Spatially resolved emission measurements directly image the wave functions of polariton states confined in the optical trap. The energy and wave function of these discrete states are well reproduced considering a particle with the polariton effective mass and using a potential $V(r)$ shown with dashed white line in Fig. 2(a).

As the excitation power is increased, the polariton population in the trap increases [see the abrupt increase of emission intensity in Fig. 2(c)] and polariton condensation is obtained on the lowest energy state of the trap [see Fig. 2(b)].

Depending on the relative size of the excitation spot and the microcavity, we can generate 0D polariton condensates which either strongly overlap with the excitonic reservoir (in small micropillars) or which are spatially separated from it (in large micropillars or in optical traps). This allows changing the relative strength of polariton-polariton and polariton-reservoir interactions.

Figure 3(a) compares the blueshift measured in micropillars of various sizes and in the optical trap. When the size of the micropillar is comparable to that of the excitation spot (as is the case in the 3.2 μm micropillar), the polariton condensate cannot spatially separate from the excitonic reservoir. A strong blueshift is measured, but the relative strength of polariton-polariton interaction and polariton-reservoir interaction cannot be quantified. Using a microwire or a micropillar much larger than the excitation spot allows the generation of a polariton condensate well-separated from the excitonic reservoir, and the role of interactions within the condensate is revealed. In these systems, the continuous blueshift observed below $3P_{\rm th}$ is

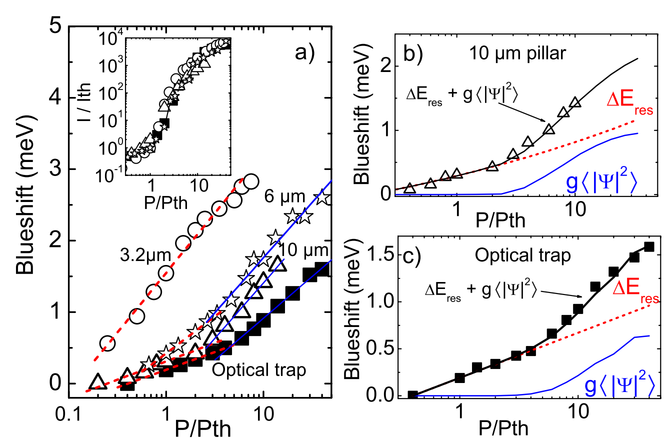

FIG. 3 (color online). (a) Blueshift measured in a 10, 6, and 3.2 μm micropillar and in the optical trap. The dashed red (solid blue) line shows the excitation range dominated by interactions with the reservoir (within the condensate). The inset shows the measured intensity in the four systems. (b) Symbols: Measured blueshift of the lowest energy polariton state in the 10 μm micropillar. Calculated blueshift induced (dashed red line) by the excitonic reservoir or (solid blue line) by interactions within the condensate deduced from the measured polariton occupancy, (black line) total calculated blueshift. (c) Same as (b) for the optical trap.

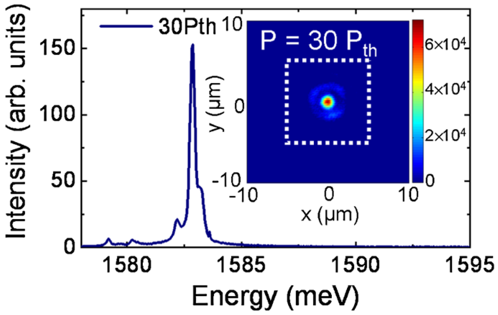

FIG. 4 (color online). Emission spectrum measured in a 10 μm square micropillar at $30P_{th}$ in the photon lasing regime. The corresponding spatial emission pattern is shown in the inset where the white dashed square shows the pillar edge.

attributed to interactions with the excitonic reservoir. This blueshift is smaller than in the 3.2 μm pillar because the spatial overlap between the polariton states and the reservoir is reduced. Above $3P_{th}$, the slope of the blueshift abruptly increases. For such high excitation powers, the polariton occupancy [27] becomes large so that polariton-polariton interactions contribute to the condensate energy renormalization. The measured blueshift is fully reproduced considering the polariton-polariton interaction term $g\langle|\psi(r)|^2\rangle \approx g\frac{I}{I_{th}}$, where $\frac{I}{I_{th}}$ is the measured polariton occupancy which can reach several thousands. The best fit of the blueshift [see Figs. 2(b) and 2(c)] is obtained for $g \approx 2\ \mu\text{eV}\cdot\mu\text{m}^2$ in the optical trap [28] and $g \approx 9\ \mu\text{eV}\cdot\mu\text{m}^2$ in the 10 μm pillar. These values are in good agreement with the theoretical one, typically $3\ \mu\text{eV}\cdot\mu\text{m}^2$, given by $3a_b^2E_x$, where a_b and E_x are, respectively, the exciton Bohr radius and binding energy [24]. Notice that the repulsive potential resulting from the high occupancy of the condensate in the trap also affects the higher energy polariton states. The energy and wave function of these states can be well reproduced taking into account the polariton-polariton interaction term found above [see, for example, Fig. 2(b)].

Another interesting spatial feature is observed at higher pumping rates in the 10 μm micropillar. As reported in Ref. [13], transition to the weak coupling and onset of photon lasing can be observed. This second threshold not only corresponds to a jump in the emission energy and an increase in the emission intensity, but also to an abrupt qualitative change in the emission pattern. As shown in Fig. 4, photon lasing occurs at the center of the pillar, where population inversion is first obtained and thus where the gain is maximum. Moreover, gain confinement in the excitation area is observed due to the change of refractive index induced by the high pumping rate. Thus, an opposite behavior is observed between the polariton condensation regime, where repulsive interactions tend to push away polaritons from the excitation area, and photon lasing which occurs tightly focused in the excitation area.

To conclude, we have shown that because of their excitonic nature polariton condensates undergo significant repulsive interaction with the reservoir of uncondensed excitons. This interaction induces dramatic changes in the polariton spatial distribution which are pushed away to the corners of a micropillar or trapped between the excitation area and the end of a microwire. These features are shown to be a polaritonic behavior since we observe the opposite effect, namely, gain confinement, when the system enters the regular photon lasing regime. Further evidence of the matter part of these Bose condensates is demonstrated for large polariton occupancy: an additional emission blueshift is the signature of polariton-polariton interactions within the condensate.

This work was partly supported by the C'Nano Ile de France contract "Sophiie2," by the ANR Contract No. PNANO-07-005 GEMINI, by the FP7 ITN "Clermont4" (235114), and by the FP7 ITN "Spin-Optronics" (237252).

*jacqueline.bloch@lpn.cnrs.fr

[1] M. H. Anderson *et al.*, Science **269**, 198 (1995).
[2] K. B. Davis *et al.*, Phys. Rev. Lett. **75**, 3969 (1995).
[3] For a review see F. Dalfovo *et al.*, Rev. Mod. Phys. **71**, 463 (1999), and references therein.
[4] D. M. Stamper-Kurn *et al.*, Phys. Rev. Lett. **83**, 2876 (1999).
[5] A. J. Leggett, Rev. Mod. Phys. **71**, S318 (1999).
[6] Gordon Baym and C. J. Pethick, Phys. Rev. Lett. **76**, 6 (1996).
[7] L. V. Hau *et al.*, Phys. Rev. A **58**, R54 (1998).
[8] J. Kasprzak *et al.*, Nature (London) **443**, 409 (2006).
[9] R. Balili *et al.*, Science **316**, 1007 (2007).
[10] C. W. Lai *et al.*, Nature (London) **450**, 529 (2007).
[11] S. Christopoulos *et al.*, Phys. Rev. Lett. **98**, 126405 (2007).
[12] G. Christmann *et al.*, Appl. Phys. Lett. **93**, 051102 (2008).
[13] D. Bajoni *et al.*, Phys. Rev. Lett. **100**, 047401 (2008).
[14] E. Wertz *et al.*, Nature Phys. **6**, 860 (2010).
[15] E. A. Cerda-Méndez *et al.*, Phys. Rev. Lett. **105**, 116402 (2010).
[16] F. Tassone *et al.*, Phys. Rev. B **56**, 7554 (1997).
[17] F. Tassone and Y. Yamamoto, Phys. Rev. B **59**, 10 830 (1999).
[18] C. Ciuti *et al.*, Phys. Rev. B **62**, R4825 (2000).
[19] S. Utsunomiya *et al.*, Nature Phys. **4**, 700 (2008).
[20] A. Amo *et al.*, Nature (London) **457**, 291 (2009).
[21] A. Amo *et al.*, Nature Phys. **5**, 805 (2009).
[22] L. Pitaevskii and S. Stringari, *Bose-Einstein Condensation* (Oxford Science Publications, Oxford, U.K., 2003).
[23] M. M. Glazov *et al.*, Phys. Rev. B **80**, 155306 (2009).
[24] C. Ciuti *et al.*, Phys. Rev. B **58**, 7926 (1998).
[25] D. Sanvitto *et al.*, Phys. Rev. B **80**, 045301 (2009).
[26] See supplemental material at http://link.aps.org/supplemental/10.1103/PhysRevLett.106.126401 for additional measurements on single micropillars.
[27] We estimate that the mean occupancy at threshold is close to unity.
[28] In the case of the optical trap, we also consider the interaction term $g\langle|\psi_3(r)|^2\rangle$ induced by the third polariton state, which is also highly populated.